Engineering Aspects of the Disposal of Radioactive Wastes from the Peacetime Applications of Nuclear Technology

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One watches with an appreciation akin to awe—and not entirely unmixed with grave concern—the growing engineering achievements in meeting the ever-deepening problem of disposal of radioactive wastes. This paper will bring reassurance to all of us in public health.

The handling and disposal of radioactive wastes is a general problem the thread of which runs through the complete fabric of peaceful nuclear energy operations. In the peacetime, day-to-day application of the benefits of nuclear energy the disposal of wastes potentially represents the major "nonbeneficial" effect on the public and its resources. Therefore, it is probably this segment of the operations that is of greatest direct interest to the public health profession. Waste materials in either gaseous, liquid, or solid form are evolved in essentially all operations associated with nuclear energy facilities beginning with mining of ore, through feed material production, reactor operation, and chemical reprocessing of reactor fuels. Because of the nature and characteristics of the radioactivity involved, its ability to cause damage to human tissue, and its potential danger as an environmental contaminant, the safe handling and final disposal of nuclear energy wastes are integral and important aspects of these operations. This importance is attested to by the efforts expended in the atomic energy program to date. More money probably has been spent, and more scientific and technological effort concentrated, on facilities, operations, and research and development with regard to this industrial waste than on any other industrial contaminant we have known.

It has been said a number of times that the widespread peaceful and beneficial application of nuclear technology will depend to a considerable degree on our ability to find practical solutions to problems of waste handling and disposal. While it reasonably can be argued that no industry can be considered a mature segment of our economy unless and until it handles and disposes of its wastes in an acceptable manner, there is sufficient basis for the belief that the nuclear energy industry can develop in a rational way without being "bottlenecked" by its wastes. This conviction should not, however, carry the implication that specific answers are immediately available. Much research, development, pilot-plant testing, and field evaluation have yet to be done before

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firm engineering conclusions will be possible for all situations.

It is important to emphasize early that the disposal of radioactive wastes, from an engineering and environmental standpoint particularly, cannot be considered as a single problem with a single, best solution. The great variation in the characteristics of the waste products from various processes and operations, including half-life, chemical state and concentration, physical nature, the quantities of materials involved, and the specific location of the nuclear facility are all important in assessing the significance of the hazard and in establishing engineering design criteria for their handling and disposal.

One of the major objectives of our engineering in this field is control over the radiation hazard from these wastes. Obviously, this involves control over the movement or mobility in the environment of the waste products themselves. This introduces the two basic waste disposal concepts that are applicable to the waste problem in its broadest sense. The radioactive materials may be permanently confined or isolated within restricted areas, away from people and their resources. This is the concept of "concentrate and contain." On the other hand, the radioactivity may be irreversibly reduced to safe levels by dilution in nature. This is the concept of "dilute and disperse." For example, with suitable environmental conditions certain types of laboratory liquid wastes in which the concentration of radioactivity is only a few times greater than drinking water standards may be disposed of under the latter concept. At the other extreme highly active liquid wastes originating from the chemical processing of irradiated fuels must be handled under the former philosophy. For all practical purposes the waste materials evolving from the chemical processing of irradiated fuels contain, by far, the greatest concentrations and total quantities of radioactivity and constitute the bulk of the long-term technological problem of waste disposal. It should be emphasized, however, that the fact that wastes containing smaller quantities of radioactivity may be amenable to direct dispersal in the environment makes it inherently important to control such operations carefully to assure that the safe capacity of the environment is not exceeded.

So much for background and philosophy. Now, what specifically, are these radioactive wastes we are concerned with? How do we handle them at the present time and what are some considerations for the future?

Characteristics of gaseous or airborne particulate wastes vary widely depending on the nature of the operation from which they originate.[1] In gaseous form they may range from rare gases, difficult to remove, such as Argon (A^{41}) from air-cooled reactors, to highly corrosive gases, such as hydrogen fluoride (HF) from chemical and metallurgical processes. Particulate materials (aerosols) may be organic or inorganic and range in size from less than 0.05 microns to 20 microns. The smaller particles originate from metallurgical fumes caused by oxidation or vaporization. The larger particles may be acid mist droplets which are low in specific gravity and may remain suspended in air or gas streams for longer periods.

An outstanding feature of air cleaning requirements for many nuclear energy operations results from the extremely small permissible concentrations of various nuclides in the atmosphere. Often removal efficiencies of the order of 99.9 per cent or greater for particles less than one micron in diameter are necessary. These criteria are much more stringent than heretofore encountered in industrial hygiene engineering, and in the early days of the industry, could not be met by the general industrial dust control equipment then available. This situation was met by a research and development program that encompassed fundamental studies on the characteristics and behavior of aerosols, the design of specialized equipment, and the evaluation of the dispersion of these materials into the atmosphere. As a result of this program such air cleaning units as high-efficiency filters capable routinely of removing 99.95 per cent of particles 0.3 microns in diameter were developed and put into commercial production. A packed tower unit which utilized the chemical reaction between iodine and silver nitrate and capable of removing I^{131} from gas streams with efficiencies greater than 99.99 per cent was designed and put into successful operation and other special air cleaning devices were developed. The principles of atmospheric diffusion and dispersion were quantitatively evaluated to the point where these factors could be utilized in the over-all design of gaseous waste treatment facilities.

Some aspects of future air cleaning problems which require further technical effort include high-efficiency filtration at temperatures above 1,000°–1,500° F and, if very short-time cooled fuels are processed, even more efficient removal of iodine. The question of the biological role of a discrete, individual radioactive particle is still under active consideration and its resolution might well influence the performance criteria of engineering facilities designed to control particulate effluents. Nevertheless, for the most part, handling and disposal of gaseous wastes from nuclear facilities is amenable to practical engineering control.

Solid radioactive wastes, such as nonusable contaminated equipment, nonrecoverable scrap, and contaminated trash produced in all operations do not constitute a serious technical problem. However, if inadequate provisions are made for their proper handling and disposal they could be a distinct nuisance and, under certain circumstances, even a hazard. The levels of radioactivity associated with solid wastes may vary from a few times background to quantities requiring substantial shielding or remote handling. The engineering of systems for handling and disposal of solid wastes has been relatively simple. Burial of such wastes under known, controlled conditions and, in specific instances, disposal at sea have successfully and safely handled the problem.

Established burial grounds for solid radioactive wastes exist only at large atomic energy production and development sites, such as Oak Ridge, Idaho, Savannah River, Hanford, and Los Alamos. These facilities are in isolated areas with detailed geology and hydrology generally favorable to burial ground location. Within AEC, the operating establishments other than those noted above usually consist of relatively small areas and are in or near densely populated sections with perhaps less favorable geology and hydrology. In these cases the general procedure is not to dispose of wastes on site but to ship to one of the established burial grounds for final disposition.

In this latter connection we are confronted with the problem of locating another suitable burial ground (or grounds) to facilitate and reduce the cost of these operations. This situation is most pressing in the northeastern United States where, at present, the only available disposal sites are Oak Ridge and the Atlantic Ocean. It is generally felt that solid waste disposal facilities should not be indiscriminately scattered around the country. (The proposed AEC regulations on standards for protection against radiation do permit "on-site" burial of very small quantities of radioactivity, however.) Accordingly, AEC staff is now engaged in investigations directed toward the establishment of a solid waste handling and disposal facility to service the northeastern United States. How such a facility will be operated has not yet been decided, but in so far as technically and administratively feasible commercial participation will be encouraged.

To date relatively small quantities of radioactivity (estimated in the range of hundreds of curies, with the major contributions coming from Brookhaven National Laboratory, Bettis Field, and the University of California Radiation Laboratory) have been disposed of at sea. In these instances this has been a safe, practical method of disposal. However, among other things, the costs of sea disposal operations, including shipping to port, etc., appear to preclude the widespread use in this country of this method over burial.

Incineration of combustible, solid wastes to reduce volume and facilitate ultimate disposal has been practiced to a limited extent. The engineering and economic advantages of incineration over other approaches, including compressing, baling, and direct burial have not been demonstrated but concentration by burning may be practical in specific circumstances. A special unit to burn 30 pounds per hour has been developed by the U. S. Bureau of Mines and is now being put into operation at the Harvard Air Cleaning Laboratory in connection with final development of a suitable air cleaning component. Where quantities of activity associated with solid wastes are low the use of a "solid dilution" approach, i.e., disposal into a municipal or institutional incinerator may be acceptable. This problem is now being considered in some detail by a special subcommittee of the National Committee on Radiation Protection of the U. S. Bureau of Standards. An NBS handbook on this subject is the objective of this subcommittee.

Liquid radioactive wastes are evolved in all nuclear energy operations from laboratory research to full-scale production. As previously indicated, it is important to differentiate between what we call a "high-volume, low-level" waste, for example, the contaminated laundry waste which may contain say a few microcuries of radioactivity per gallon, and a "low-volume, high-level" waste resulting from chemical processing of nuclear reactor fuels which may contain up to 1,800 or more curies per gallon. Although both categories are radioactive wastes and both are liquid the similarity ends right there. The engineering problems of handling and disposing of these two categories are entirely different.

Liquid wastes with low concentrations of radioactivity originate in laboratory operations where relatively small quantities of radioactive materials are involved—ore and feed material processing, the normal operation of essentially all reactors, particularly water-cooled types, and the routine operation of chemical processing plants. These low-activity wastes, under proper environmental conditions, are susceptible to either direct disposal to nature or to disposal following minimum treatment. Treatment processes used include coprecipitation, ion-exchange, biological systems similar to sewage treatment methods and others. Because of their relatively high volume (on the order of millions of gallons per day) total costs for treatment may be substantial. Therefore, to the extent that it is absolutely safe, maximum use is made of dilution factors that may be available in the environment and that can be assessed quantitatively. This points up the importance of proper site selection for nuclear energy facilities and the necessity for quantitative data concerning the environment.

High-activity liquid wastes associated with the chemical processing of reactor fuels, as already indicated, constitute the bulk of the engineering problem of disposal of radioactive wastes. It should be pointed out clearly that these wastes do not come directly from the reactors themselves, although under the improbable conditions of reactor malfunction some high-activity waste material may result. In the future such wastes also may be associated with certain types of homogeneous reactor sites to the extent that continuous fuel processing right at the reactor is envisaged. At the present time, however (and very likely for the immediate future), chemical processing plants are essentially the sole source of the wastes. Production scale chemical plants are presently located at Savannah River, Hanford, and Idaho.

Chemical processing of reactor fuels is done to separate and recover unfissioned or unburned fuel from the desired product and the wastes. At the present time, this means for the most part separating uranium, plutonium, and fission products. The fission products which constitute the "hot" component of the wastes are radioactive elements ranging in mass number from 70 to 162. From an environmental standpoint the more troublesome of these elements include strontium, cesium, and others in the rare earth series. In the solid fuel element prior to separation (which is today generally an aqueous chemical process) the concentration of fission products ranges from 100 to perhaps 1,000 ppm. In the course of processing the fission product concentration is considerably diluted by solvents, water, and other solids. The resulting liquid wastes streams are, therefore, quite dilute as far as mass concentration of fission products is concerned. However, as previously stated, because of the high specific activity of these elements, these wastes may contain quantities of radioactivity up to several hundreds of curies per gallon. The effective life of this radioactivity may be measured in hundreds of years. These wastes also may generate heat to the extent of 10–50 Btu per gallon per hour.

The quantity of high-level wastes generated depends largely on the chemical and metallurgical characteristics of the fuel being processed and the specific nature of the chemical process involved. It may range from 0.1 gallons to 5.0 gallons per gram of uranium processed. On a total volume basis it is estimated that we have to consider waste production rates of the order of a few tens of millions of gallons per year. The accompanying table gives the characteristics of a high-activity waste from processing materials testing reactor fuel elements.

First Cycle Waste Characteristics
MTR Fuel Element Processing

Specific Volume, l/Kg U.	413
Aluminum Nitrate, *M*	2.1
Nitric Acid, *M*	0.9
Sodium Nitrate, *M*	0.1
Mercuric Nitrate, *M*	0.002
Density, g/cc	1.35
Total Radioactivity, Curies/gallon	1.9×10^3
Heat Generation, Btu/(gal) (hr)	25 ($\frac{1}{2}\beta$, $\frac{1}{2}\gamma$)

Notes: Calculations based on following conditions:
1. 20 per cent burn-up.
2. 120 day cooling.
3. Prior to jetting from evaporator.

From the viewpoint of the sanitary engineer it is perhaps misleading to apply the term "disposal" to current methods of handling highly radioactive liquid wastes. With only minor exceptions these wastes are not "disposed of" but are stored in specially designed tanks. Since the effective life of the fission products constituting the wastes may be measured in terms of hundreds of years, it is apparent that tank storage is not a permanent, long-term answer to the disposal problem. The latter point is accentuated by the fact that the capital cost of tank storage varies from about 30 cents to roughly $2 per gallon capacity.

What then, are the possibilities for disposing of the high-level wastes? First, based on various estimates on the growth of nuclear energy in this country one can calculate the total cumulative quantity of radioactivity to be disposed of at given times in the future. Depending on whose nuclear energy growth estimates are used the radioactivity accumulations range from about $3 \times 10^9 - 2 \times 10^{10}$ curies in 1965 to about $4 \times 10^{11} - 1 \times 10^{12}$ in the year 2000. Now, when one considers the generally extremely low maximum permissible concentrations of radioactivity in air and water it becomes apparent that there is not enough dilution available in nature to enable any practical, continuing dispersal of these wastes into the environment. The application of the dilute and disperse philosophy does not appear to be a possibility.

A possible exception, but somewhat academic at the present time is disposal at sea.[2] Some oceanographers have indicated that based on general knowledge of the behavior of the ocean deeps (12,000–15,000 feet depths) and marine biology, it appears that substantial quantities of radioactivity may be disposed at these depths safely. They also indicate that much detailed oceanographic work is required before actual sea disposal criteria could be established. This present lack of detailed oceanographic information coupled with the complete loss of control of the material once it is disposed, the complex engineering problems, and high estimated costs involved in handling, transporting, and actual placing of these wastes in the ocean deeps leads once to the conclusion that sea disposal is probably a secondary possibility at this time.

Several practical approaches to ultimate, safe disposal of high-level wastes appear possible.[3] These possibilities are briefly described below. Work on these approaches is being carried out at a number of AEC installations.

Fixation in Inert Media—The objective is to fix the radioactive waste material, i.e., the actual fission products, in an inert solid carrier so that the possibility of migration of the radioactivity into the environment is eliminated or reduced to acceptable and safe limits. The carrier containing the radioactive material could then be permanently stored or buried in selected locations without deleterious effect on man or his environment. Fixation on clay, incorporation in feldspars, conversion to oxide elutriation of oxide fixation of elutriant are examples of systems under development.

Special Separation of Specific Isotopes—Because of the particular radiotoxicity and long half-life of Sr^{90} (25 years) and Cs^{137} (33 years), the removal and separate fixation and handling of these two isotopes would substantially reduce the effective life of the remaining material and facilitate its final disposal. With Cs and Sr removed the possibilities of safe disposal into the environment under controlled conditions are greatly increased. The economic utilization of the Cs, particularly, is an added incentive to such separations. Since Cs has fairly energetic β and γ emissions and a 33-year half-life it is a useful radiation source.

Direct Discharge to Selected Geologic Formations—Preliminary evaluations indicate the possible technical feasibility of direct disposal of highly radioactive liquids into the ground following somewhat similar practices in other industries, but taking into account the unique characteristics of radioactive wastes. It may be practical to dispose of the wastes underground in some cases without any treatment, into such formations as: (1) spaces prepared by dissolution in salt beds or salt domes; (2) deep basins (5,000–15,000 feet in depth) containing connate brines and with no hydraulic or hydrologic connection to potable waters or other potentially valuable natural resources; and (3) special excavations in selected shale formations.

There are a number of engineering problems that must be solved before final engineering of a prototype installation utilizing the above schemes is possible. In the fixation schemes the requirement for equipment to withstand highly corrosive media, to control highly active aerosols which are evolved, and which can be operated remotely with absolutely minimum maintenance is no small order. Progress along these lines is being made, however. In the separation of specific nuclides the outstanding requirement is the high degree of separation required in order to handle the remaining material essentially as a low level waste. Decontamination factors of the order of 10^6 are necessary, i.e., 99.9999 per cent removal of Cs and Sr. Here, too, progress is being made. In the direct disposal systems questions relating to the physical and chemical reactions between the wastes and the formation material, control of thermal heat due to radioactive decay, and potential transport problems have to be answered. Work along these lines is now in its beginning stages.

One aspect of the over-all subject of waste disposal which deserves special note is that of site selection for nuclear energy facilities.[4] It should be emphasized here that it is essential in any evaluation of, or attack on, the problem of wastes to integrate fully the varying conditions of site. This may seem like an obvious statement hardly requiring emphasis; nevertheless, it is surprising to observe the past instances in other industries in which such integration was not properly carried out and resulted either in increased costs or problems of environmental contamination. It is gratifying to note the positive attitude prevalent in industry today with regard to these problems. In planning for the expansion of the nuclear energy industry special emphasis should be placed on this point of site selection. This important consideration accounts for our continuing, close, day-to-day working relationship with the meteorologist, geologist, hydrologist, and associated disciplines.

Specific note must also be made of the administrative aspects of the operation of the nuclear energy industry, including control, regulations, and public relations. The various disciplines in public health, through experience, are particularly cognizant of the importance of these considerations. They also recognize the very close tie-in between these factors and waste handling and disposal, and the vital necessity for a firm base of scientific and engineering information on which they can establish rational administrative criteria and procedures.

Changing technology is a characteristic of the nuclear energy business. New reactors, new fuel elements, new chemical processes, and new uses of radiation may have a profound effect on the nature of the waste problem and its solution. In the future it will become perhaps even more important to maintain a close working relationship between the nuclear engineer, the chemical engineer, the sanitary engineer, and the environmentalist. To my knowledge no other industry has faced the challenge of the diverse problems in waste disposal that is in front of the nuclear energy industry. But we who are working on this fascinating and challenging problem look forward to the future of our industry with courage and confidence. In this scene the public health profession has its part to play and its contribution to make.

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